Study of the Isobars Cd¹¹³^m and In^{113m}[†]

E. DER MATEOSIAN AND M. GOLDHABER Brookhaven National Laboratory, Upton, New York 11973 (Received 11 April 1969)

The decay schemes of Cd^{113m} and In^{113m} have been studied. An isomeric transition from Cd^{113m} to the ground state, previously reported by the authors, has been further investigated. An upper limit has been placed on the probability for K capture in In^{ii₃₇} (log $f(z)$). The conclusion is drawn that Cd¹¹³ is the heavier member of the Cd¹¹³-In¹¹³ isobaric pair, with an available β -decay energy of 316 keV.

I. INTRODUCTION

THE question of which of the two "stable" adjacent **L** isobars Cd¹¹³ and In¹¹³ undergoes β decay has been the subject of investigation for some time.^{$1-7$} A report³ that L capture may have been seen in In¹¹³ was later contradicted' and lower limits of the order of 10'4—10" yr were set on K capture,⁴ L capture,⁵ and positron emission⁴ in In¹¹³, and β decay^{4,6,7} in Cd¹¹³.

An isomeric state of Cd¹¹³ with a half-life of 5.1 yr was reported by Carss, Gum, and Pool.⁸ They observed β rays with an end-point energy of 500 keV accompanied by a 10% x-ray branch. Later, the halflife of this isomer was reported by Wahl and others' to be $\tau_{1/2}$ = 14 yr. Some time ago we showed¹⁰ that most of the 10% x-ray branch was probably due to an impurity (Cd^{109m}) in the Cd^{113m} sample, but that there existed a weak (0.1%) 265-keV γ -ray branch which was interpreted as an isomeric transition from the excited state to the ground state in Cd¹¹³. The β -ray end-point energy was measured by introducing a small amount of Cd^{113m} into a melt from which a $NaI(Tl)$ scintillating crystal was grown, and using this crystal as a detector with essentially 4π geometry for the zerothickness source of Cd^{113m} β rays dispersed throughout the crystal. The end-point value obtained¹¹ was 0.58 MeV, in agreement with a measurement reported by

-
-
- ⁵ Heintze and Haxel, Eur. Sci. Notes (Office of Naval Research)
8, 110 (1954); J. Heintze, Z. Naturforsch. 10a, 77 (1955).
⁶ M. I. Kalkstein, Ph.D. thesis, University of Chicago, 1952 (unpublished) .
-
- 'H. Selig, Ph.D. thesis, Carnegie Institute of Technology, 1956 (unpublished) . ' W. L. Carss, G. R. Gum, and M. L. Pool, Phys. Rev. 80, 1028 (1950);J. R. Gum, L. E. Thompson, and M. L. Pool, ibid.
- 76, A184 (1949). ⁹ A. C. Wahl, J. Inorg. Nucl. Chem. 10, ¹ (1959);K. F. Flynn, L. E. Glendenin, and E. P. Steinberg, Nucl. Sci. Eng. 22, 416
- (1965); A. Veres and M. Ozhdyani, Yadern. Fiz. 3, 798 (1966)
[English transl.: Soviet J. Nucl. Phys. 3, 587 (1966)].
¹⁰ E. der Mateosian and M. Goldhaber, Bull. Am. Phys. Soc.
1, 207 (1956).

186

Cassidy¹² (0.59 MeV). Attempts to prove that the 265 γ ray was indeed associated with Cd^{113m} and was the isomeric transition and not a transition following a β branch were, however, inconclusive because of the weakness of the source and the relatively insensitive techniques available at that time.

Recently, taking advantage of new and more sensitive techniques, the authors have reinvestigated the original Cd¹¹³ source, which was made in 1955, and have succeeded in showing that both the β and γ radiations follow Cd chemistry, and that the 263.7 ± 0.3 -keV γ ray is indeed the isomeric transition to the ground state in Cd¹¹³. These findings establish Cd¹¹³ as the heavier (potentially β -unstable) member of the isobaric pair. An upper limit has also been set on a K capture branch from the isomeric state In^{113m} (100 min) to the ground state of $Cd¹¹³$.

II. SOURCE PREPARATION AND CHEMICAL ANALYSIS

Two independently prepared sources of Cd^{113m} were used in these investigations. One source¹³ consisted of a few mg of enriched Cd¹¹² in the form of CdO which was irradiated in the Brookhaven Graphite Research Re $actor^{14}$ for 2 months and then was put aside to allow impurities to decay. Samples of Cd enriched in isotopes other than Cd¹¹² were also irradiated at this time and were helpful in assigning the activity to Cd^{113m} . β -ray studies were made with the source several years after irradiation, while x-ray and γ -ray studies were made as late as 12 yr after irradiation. A second and stronger source was made later by irradiating in the Materials Testing Reactor¹⁵ (MTR) for 3 months a 12-mg sample of CdO enriched in Cd¹¹². Both samples underwent chemical treatment to remove impurities. The CdO was dissolved¹⁶ by wet ashing with concen-

f Work performed under the auspices of the U. S. Atomic Energy Commission.
¹ E. Zingg, Helv. Phys. Acta 13, 219 (1940).

² L. T. Rusinov and J. H. Igelnitsky, Compt. Rend. USSR 47,

333 (1945); 49, 343 (1945).

³ S. G. Cohen, Nature 167, 779 (1951).

⁴ E. A. Martell and W. F. Libby, Phys. Rev. 80, 977 (1950).

¹¹ E. der Mateosian, Phys. Rev. 95, 646A (1954).

¹² J. M. Cassidy, Phys. Rev. 83, 483 (1951).

¹³ Enriched isotope samples were obtained from Stable Isotope Sales, Oak Ridge National Laboratory, Oak Ridge, Tenn. '4 Brookhaven Graphite Research Reactor, Brookhaven Na-

tional Laboratory, Upton, N. Y.
- ¹⁵ Materials Testing Reactor, Idaho Nuclear Corp., Idaho

Falls, Id.

¹⁶ All chemical procedures were performed by E. F. Norton of the Chemistry Department, Brookhaven National Laboratory, Upton, N. Y. 1285

FIG. 1. X-ray spectra observed in Cd^{113m} sources differing in physical and chemical structure. Spectrum a (solid dots) was obtained from an \sim 1-mg CdO sample before chemical purification. Spectrum b (squares) was obtained after the CdO was dissolved and the Cd was precipitated as cadmium 8-hydroxy quinolate. Spectrum c (circles) was obtained after the cadmium $\&$ -hydroxy quinolate was converted back to the oxide by slow
ignition. The difference between curves a and c is due to the fact that after the second precipitation the CdO was more widely dispersed than in the original sample, thus giving rise to fewer x rays through external ionization.

trated nitric acid and 72% perchloric acid. An ionexchange separation on Dowex-1 was performed; the cadmium was reprecipitated as cadmium 8-hydroxy quinolate and mounted for counting. Radiations from these samples were investigated before and after chernical purification.

A. Experimental Results

$1. \text{Cd}^{113m}$

Investigation of the Cd^{113m} samples after they had been allowed to cool for a minimum of 5 yr showed that the predominant radiations emitted by the samples were β rays with an end-point energy close to the published value¹¹ of 0.58 MeV. In addition, a low-intensity γ ray was observed with a lithium-drifted germanium detector and its energy was determined to be 263.7 ± 0.3 keV. Partially resolved K x rays of Ag, Cd, and In were detected with a lithium-drifted silicon detector with a resolution of 0.47 keV at 6.4 keV (Fe K_{α} line). Although the relative intensity of the γ and β rays remained constant before and after chemical purification of the samples, the relative intensity of the cadmium K x rays changed by factors as great as 5. The data shown in Fig. 1 indicate that this fluctuation in the Cd K x-ray intensity was due to a sample-dependent contribution to the Cd K x rays resulting from a β -rayinduced external ionization of Cd in the sample. The x radiations observed from a sample of Cd^{113m} , in the chemical form of CdO, are shown in curve a of Fig. 1. This sample then underwent the chemical procedure described above and was precipitated as cadmium 8-hydroxy quinolate, from which spectrum b was obtained. The amount of Cd in both samples was the same; the dramatic change in intensity of the Cd K x rays was due to the "dilution" of the Cd atoms by the hydroxy quinolate radicals. Finally, the sample was reconverted to CdO by a slow ignition, and spectrum c was obtained showing an increase in the Cd \overline{K} x-ray intensity. These results led to the conclusion that the change in Cd K x-ray intensity was due to a sample-dependent effect and not due to the presence of other radioactive impurities emitting Cd K x rays. The spectrum in Fig. 2 was observed with a 5-yr-old sample of Cd^{113m} which was put into a water solution in order to reduce the β -ray ionization of the Cd to a negligible amount. The presence of an 88-keV γ ray was detected in the photon spectrum emitted by this sample, and this suggested that the Ag K rays in Fig. 2 are due to a Cd¹⁰⁹ impurity in the sample. The In K rays are of an intensity compatible with the assumption that they result from autoionization accompanying the β decay of the Cd^{113m} isomer.

The relative intensities of the β and γ radiations of Cd^{113m} were measured in two ways. The radiations of a

FIG. 2. X-ray spectra emitted by a Cd^{113m} source observed with a lithium-drifted silicon detector. The resolution of this detector was 275 eV at 6.4 keV (Fe $K\alpha_1$ line). The Ag K x rays are ascribed to a Cd¹⁰⁹ impurity. The In K x rays are most probably due to auto-ionization accompanying β rays emitted by the Cd¹¹³ In the Cd E x rays are interpreted to be due to conversion isomer. The Cd E x rays are interpreted to be due to conversion of the 263.7-keV isomeric transitions.

	Contract Contract Theory				
	E4	Μ4	E5	Μ5	Experimental value
α_K	0.62	2.5	2.5	11	
$\tau_{1/2}$ (sec)	6×10^4	2.6×10^6	9.1×10^{10}	3.1×10^{11}	1.9×10^{12}

TABLE I. Comparison of theoretically predicted and experimentally determined K-conversion coefficients and half-lives for a 263.7 -keV transition in Cd¹¹³.

thin Cd^{113m} source were compared with the radiations of a thin T1²⁰⁴ source. T1²⁰⁴ is chiefly a β emitter which has a 2% K-capture branch. It emits no γ rays in its decay. The Cd^{113m} and Tl²⁰⁴ β -ray intensities were compared with a plastic scintillator in 2π geometry; the intensity of the 263.7-keV γ ray of Cd^{113m} was compared with the intensity of the Hg $K \times$ rays emitted by the T^{204} source with a 3-in. $NaI(Tl)$ detector. The value obtained for the intensity ratio of the 263.7-keV γ ray to the β rays from Cd^{113m} was 2.4×10⁻⁴. A second determination of this ratio was made with a lithiumdrifted germanium detector which was used in a known geometry. The β -ray intensity was again measured with a plastic scintillator used in 2π geometry. A ratio of 2.2×10^{-4} was obtained. The average of these values, 2.3×10^{-4} , was adopted for the γ -ray branch in Cd^{113m}.

The best value for the K conversion of the 263.7-keV γ ray (α_K , expt) was calculated from the relative intensities of Cd K x rays and the 263.7-keV γ ray. This was obtained with the liquid source of Cd^{113m} by measuring the relative intensities of the total K x rays and γ rays (see Fig. 3) with a 3-in. integral-line¹⁷ NaI(Tl) detector, and then applying a correction to determine the Cd K x-ray component of the x rays from the data taken with the same liquid source by means of a

F I^T I'IG. 3. Energy spectrum of photons emitted by a Cd¹¹³ sample and detected with a 3×3-in. NaI(Tl) scintillation spectrometer

lithium-drifted Si detector (see Fig. 2). The value obtained for (α_K, expt) is 3 \pm 0.5.

A comparison is made in Table I of (α_K, expt) with calculated values¹⁸ of α_K for relevant multipolarities of transitions of this energy. Agreement exists between (α_K, expt) and both $\alpha_K(M4)$ and $\alpha_K(E5)$. The halflife for the electromagnetic transition in Cd^{113m} can be calculated from the value for the γ branch (2.3 \times 10⁻⁴) and the half-life for the isomeric state Cd^{113m} (14 yr). The computed half-life $(6 \times 10^4 \text{ yr})$ is compatible with The computed half-life $(6 \times 10^4 \text{ yr})$ is compatible with an E5 transition,¹⁹ which would be expected from a spin-parity assignment of $\frac{11}{2}$ to the isomeric level in Cd¹¹³, in keeping with shell-model predictions for this mass region.

Coincidences and anticoincidences between the β rays and the 263.7-keV γ rays were investigated by sand-

FIG. 4. β - γ coincidence and anticoincidence spectra of Cd^{113m}. Both spectra are photon spectra obtained in coincidence and anticoincidence conditions with the β rays emitted by a Cd¹¹³ sample. As seen in the accompanying sketch, the geometry and efficiency for detecting β rays was very close to 4π and 100% , so that the efficiencies with which the photons were detected were the same under the two conditions of coincidence and anticoincidence. The prominent peak in the anticoincidence run is due to the 263.7-keV γ ray.

^{&#}x27;7 Harshaw Chemical Co.

¹⁸ L. A. Sliv and I. M. Band, in Alpha-, Beta-, and Gamma-Ray S*pectroscopy*, edited by Kai Siegbahn (North-Holland Publishing
Co., Amsterdam, 1965). Co., Amsterdam, 1965).
¹⁹ S. A. Moszkowski, in Alpha-, Beta-, and Gamma-Ray Spec-

troscopy, edited by Kai Siegbahn (North-Holland Publishing Co., Amsterdam, 1965); M. Goldhaber and A. W. Sunyar, in Alpha-, Beta-, and Gamma-Ray Spectroscopy, edited by Kai Siegbahn (North-Holland Publishing Co., Amsterdam

FIG. 5. Decay schemes of Cd^{113m} -In^{113m}.

wiching a CdO sample between two small scintillating pastic detectors $(0.5 \times 0.5 \times 0.25)$ in.) mounted on a photomultiplier tube and placing this combination source and detector close to a lithium-drifted Ge detector. This arrangement allowed the β rays to be counted in 4π geometry and with virtually 100% efficiency, so that both coincident and anticoincident runs were made under highly effective conditions. No $\beta-\gamma$ coincidences were observed; the 263.7-keV γ ray was wellresolved above background (see Fig. 4) in the anticoincident runs.

2. In^{113}

Our study of the Cd¹¹³-In¹¹³ isobars was extended to a search for evidence of K orbital-electron capture leading from the first excited state of $In¹¹³$ to the ground state of Cd¹¹³. The first excited state in $In¹¹³$ is an isomeric state with a half-life of 100 min. This activity was obtained conveniently by milking a sample of 115-day Sn¹¹³, which was formed¹³ by irradiating enriched Sn¹¹² in the Brookhaven Graphite Research Reactor.¹⁴ The radioactive sample of SnO_2 was put into solution¹⁶ and loaded into a column of Dowex-1, $X-8$, and samples of In^{113m} were obtained by elution with $1M$ HCl.

The x-ray spectrum of In^{113m} was investigated with the lithium-drifted Si detector. No Cd x rays were seen and an upper limit was established of 0.001 times the intensity of In K x rays present from the conversion of the 393-keV isomeric transition in In¹¹³. This establishes a lower limit of log $ft \sim 6$ for K capture in In¹¹³.

III. DISCUSSION AND SUMMARY

These experimental results are summarized in Fig. 5. The presence of Cd x rays and the results of β - γ coincidence and anticoincidence experiments allow us to assign the 263.7-keV γ ray to the isomeric transition in Cd¹¹³. A spin and parity of $\frac{11}{2}$ has been assigned to the first excited state in Cd¹¹³ from considerations of energylevel systematics, the details of the β decay, and the lifetime of the isomeric transition. The characterization of this transition as $E5$ is supported by the agreement of the predicted K -conversion coefficient¹⁸ 2.5 and the measured value 3. The half-life for the isomeric transition is 6×10^4 yr, which is 20 times longer than the transition is 6×10^4 yr, which is 20 times longer than the single-particle prediction for an $E5$ transition.¹⁹ A lower limit of log $ft=6$ for the K-capture branch from In^{113m} to Cd¹¹³ is compatible with β -decay systematics.

The decay scheme in Fig. 5 indicates that $Cd¹¹³$ is the heavier member of the Cd¹¹³-In¹¹³ isobaric pair. The energy difference between these isobars (316 keV) may be compared with the value given in the 1964 Mass Tables²⁰ (297 keV). The β decay of Cd¹¹³ (ground state) to In¹¹³ has not as yet been observed. The lower limit set for the log_{fo}t value (>22) for this decay is compatible with a fourth-forbidden transition. An estimate of the $\log f_0 t$ expected for this decay may be obtained by considering the reliably known example of a fourth-forbidden transition in In¹¹⁵. The log f_ot for the β decay of $In¹¹⁵$ is 22.7. An argument can be made that the Cd¹¹³ β transition ($s_{1/2} \rightarrow g_{9/2}$) will be hindered relative to the In¹¹⁵ decay on the following grounds: The ground state of $_{48}$ Cd¹¹³ has a spin and parity of $\frac{1}{2}$ ⁺. In the singleparticle model with configuration mixing, this state may be considered to be due²¹ to the odd neutron in an $s_{1/2}$ state and the protons in a mixture of states, such as $g_{9/2}^{10}$ and $g_{9/2}^{8}p_{1/2}^{2}$, which are coupled to $J=0$. The β decay proceeds through the conversion of an $s_{1/2}$ neutron into a $g_{9/2}$ proton. If, as is believed to be the case, the $g_{9/2}$ ¹⁰ configuration is the dominant one, the β decay will be slowed down. In¹¹⁵, on the other hand, has 49 protons. It has a $\frac{9}{2}$ + ground state and can therehas 49 protons. It has a $\frac{9}{2}^+$ ground state and can there-
fore only have a $g_{9/2}^{-1}$ proton configuration.²¹ In the β decay, an $s_{1/2}$ neutron changes into a $g_{9/2}$ proton, and the transition rate can be nearly "intrinsic." On the other hand, the statistical weight factor favors the transition $\frac{1}{2} \rightarrow \frac{9}{2}$ by a factor of 5. If everything else were equal, one may expect from these considerations a $\log ft$ $>$ 22 for the Cd¹¹³ β decay, which happens to agree with the experimental limit.

ACKNOWLEDGMENTS

The authors wish to express their thanks to R. W. Stoenner and E. F. Norton for their aid with the chemical aspects of this problem.

 \sim

 20 J. H. E. Mattauch, W. Thiele, and A. H. Wapstra, Nucl. Phys. 67, 1 (1965).

 21 A. de-Shalit and M. Goldhaber, Phys. Rev. 92, 1211 (1953).